

REPORT DOCUMENTATION PAGE

AD-A221 932		TIC		1b. RESTRICTIVE MARKINGS NONE	
2b. DECLASSIFICATION/CONTINUATION NUMBER(S) NONE		3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; Distribution unlimited.		5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR-TR- 90 0638	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) SDL 0142/0149 FTR		6a. NAME OF PERFORMING ORGANIZATION Washington State University		7a. NAME OF MONITORING ORGANIZATION Air Force Office of Scientific Research	
6b. OFFICE SYMBOL (If applicable)		6c. ADDRESS (City, State, and ZIP Code) Department of Physics Pullman, WA 99164-2814		7b. ADDRESS (City, State, and ZIP Code) Building 410 Bolling AFB Washington, DC 20332-6448	
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR/NA		8b. OFFICE SYMBOL (If applicable) AFOSR/NA		9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER AFOSR 87-0081	
8c. ADDRESS (City, State, and ZIP Code) AFOSR/NA Bolling AFB DC 20332-6448		10. SOURCE OF FUNDING NUMBERS		11. TITLE (Include Security Classification) Nonlinear Material Response to Very Rapid Energy Deposition	
13a. TYPE OF REPORT Final		13b. TIME COVERED FROM 861001 TO 891231		14. DATE OF REPORT (Year, Month, Day) 900323	
15. PAGE COUNT 32		16. SUPPLEMENTARY NOTATION		17. COSATI CODES	
18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) shear measurements, phase transitions, shock waves, lattice stress, inelastic deformation, multiple pulse, laser damage, transparent materials, electron heating, lattice defects.		19. ABSTRACT (Continue on reverse if necessary and identify by block number) Results of a research effort to examine the nonlinear response of selected materials to rapid energy deposition are summarized. One part of the work focused on examining the response of brittle solids to plane shock waves. The other part of the work focused on understanding the mechanisms for laser energy deposition in transparent dielectrics. The shock wave effort has demonstrated the usefulness of making shear wave measurements for characterizing the shocked state. In fused silica, these measurements led to the finding of reversible, shear enhanced compaction and to a direct determination of stress deviators in the shocked state. The work on polycrystalline calcite represents the first study to use shear wave measurements to understand shock induced phase changes. Shear modulus decrease associated with the calcite I-II transition was observed. The subsequent increase in shear modulus at higher compressions is surprising and, in conjunction with the longitudinal measurements, leads to the possibility that a phase other than calcite III is formed under shock loading. Results		20. DISTRIBUTION/AVAILABILITY OF ABSTRACT <input type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS	
21. ABSTRACT SECURITY CLASSIFICATION UNCLASSIFIED		22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. Spencer T. Wu		22b. TELEPHONE (Include Area Code) (202) 767-6962	
22c. OFFICE SYMBOL AFOSR/NA		23. NAME OF MONITORING ORGANIZATION Air Force Office of Scientific Research		24. ADDRESS (City, State, and ZIP Code) Bolling AFB, DC 20332-6448	

18. (continued)
optical spectroscopy.

19. (continued)
of a small effort to examine the shock response of calcite single crystals reveal a complex, time-dependent response between 22-40 kbar. Continuum measurements alone are insufficient to identify the various phases.

During the course of this work, the ability to do time-resolved, optical spectroscopy in shocked solids was demonstrated. Luminescence measurements were obtained in ruby samples subjected to 100 kbar dynamic tension. Raman measurements in shocked diamond revealed frequency shifts that are consistent with calculated strain-induced changes in the phonon frequency of diamond. These spectroscopy developments represent new approaches to characterize shocked solids.

The laser effort concentrated on nonlinear interactions between high intensity pulsed laser beams and transparent solids. New experimental techniques used to measure multiphoton absorption and rapid energy deposition in wide-gap halides and silica in the pre-breakdown regime have led to hard evidence refuting the traditional avalanche model of laser-induced damage at visible wavelengths. These measurements, performed in specially purified materials, have allowed the discovery of the roles of laser-induced excitations in rapid energy deposition, leading to the conclusion that virtually all lattice heating occurs via nonlinear absorption of laser photons by multiphoton excited free electrons. These results yield an experimentally confirmed theoretical definition of intrinsic, single pulse laser damage at 532 nm wavelength in three and four photon bandgap alkali halides.

Extending this work to multiple pulse effects in the sub-threshold intensity regime, we have also formulated and experimentally verified a new theory of intrinsic bulk damage based on thermomechanical stress induced by the accumulation of multiphoton generated lattice defects.

Washington
State University

DEPARTMENT OF PHYSICS

90 05 25 039

**NONLINEAR MATERIAL RESPONSE TO
VERY RAPID ENERGY DEPOSITION**

Final Report for
AFOSR Grant 87-0081 (DOD/URI)

Contract Period: 10/01/86 - 12/31/89

Accession For	
NTIS	<input checked="" type="checkbox"/> CRA&I
	<input type="checkbox"/> TAB
	<input type="checkbox"/> Unannounced
Justification	
By	
Distribution /	
Availability Codes	
Dist	Availability for Special
A-1	

**NONLINEAR MATERIAL RESPONSE TO
VERY RAPID ENERGY DEPOSITION**

Final Report for
AFOSR Grant 87-0081 (DOD/URI)
Contract Period: 10/01/86 - 12/31/89

Principal Investigators: Y.M. Gupta and P.F. Braunlich
Department of Physics
Washington State University
Pullman, WA 99164-2814

Prepared for the
Air Force Office of Scientific Research

Project Monitor: Dr. Spencer T. Wu
AFOSR/NA
Department of the Air Force
Bolling Air Force Base
Washington, DC 20332-6260

PREFACE

This report summarizes the work carried out over a three-year period under AFOSR sponsorship. Because this effort was part of the DOD/URI program, a considerable emphasis was placed on graduate student training and acquisition of equipment that would enhance future research. A workshop was held in May, 1989 to communicate our results and to interact with personnel from the Air Force and other organizations.

Dr. Spencer Wu was the technical monitor for this effort and he is thanked for his enthusiastic interest in this work and support. This work would not have been possible without the dedicated effort of the many students and research personnel whose names are listed in Appendix B of this report. Ms. Tamera Filler deserves a special acknowledgement for her organizational skills and hard work while taking care of the many administrative and accounting tasks.

Table of Contents

I. INTRODUCTION	1
II. SUMMARY OF THE SHOCK WAVE EFFORT	2
III. SUMMARY OF THE LASER DEPOSITION EFFORT	11
IV. CONCLUDING REMARKS	19
APPENDICES	21
Appendix A: Publications and Presentations	21
Appendix B: Personnel and Equipment Support	27
Appendix C: AFOSR Workshop Program (May 1989)	29

I. INTRODUCTION

The response of a broad range of materials (ceramics including optical windows, geophysical solids, polymers, composites, etc.) to intense impulsive loading is central to many Air Force programs and applications. With rapid advances in materials processing and fabrication technology for novel materials, and the wide spectrum of applications envisioned for these materials, it is difficult to test materials under all conditions of interest. Hence, a fundamental understanding of mechanisms governing material response under impulsive loading is needed. Understanding and predicting the highly nonlinear material response for a diverse range of materials to extreme loading conditions is a challenging problem. Knowledge gained from quasi-static deformation studies is of limited value because the underlying deformation mechanisms at high strain-rates can be quite different. In the case of energy deposition from an intense laser pulse, there is the additional need to understand the fundamental processes controlling the rapid energy deposition in materials prior to the so-called damage threshold.

The overall objective of this research effort was to understand the nonlinear material response due to very rapid energy deposition in selected solids. Two different, though related aspects of the overall problem were addressed in the present work. In the first, the response of brittle materials to plane shock waves (produced by projectile impact) was examined using novel methods. The thrust of this effort was to understand the mechanisms for material deformation at very high strain-rates and stresses. In the second aspect of the research program, the detailed microscopic mechanisms that are responsible for the absorption of laser energy in transparent dielectrics and subsequent mechanical and thermodynamic response of the material were investigated.

An extended summary of the shock wave and the laser deposition efforts is presented in Sections II and III, respectively. The emphasis in these sections is on problem definition and summarizing the research accomplishments. Detailed accounts of these efforts may be seen in journal articles and/or dissertations that are listed in Appendix A. These articles and dissertations have been submitted to AFOSR. A related goal of the present work was the training of graduate students and interaction with national laboratories. Information regarding these objectives is listed in Appendices B and C, respectively.

II. SUMMARY OF THE SHOCK WAVE EFFORT

Plane shock wave experiments provide a unique method for understanding material response at very high stresses and strain-rates. The ability to make time-resolved measurements, with nanosecond resolution, under well defined strain conditions is particularly attractive for studying time-dependent processes responsible for material deformation. Despite the extensive literature on the subject, there remain two main difficulties in understanding micro-mechanical mechanisms under shock loading: (i) the inability to experimentally determine the complete stress state in the material and (ii) the lack of direct experimental information at the microscopic level. The present work addresses these long standing problems using recent experimental developments.

The primary goals of our work were to examine the response of selected materials using combined compression and shear loading, and to explore the development of time-resolved optical spectroscopy for studying high strain-rate deformation of solids. The following four tasks comprised the shock effort: (i) Shear wave measurements in shocked fused silica to understand structural changes accompanying nonlinear elastic deformation in this material, (ii) Use of combined compression and shear loading in shocked calcium carbonate (Carrara marble) to understand structural phase transitions and to examine the interrelationship between material strength and phase transitions, (iii) To explore the development of time-resolved spectroscopic methods to understand shock induced deformation in single crystals at the microscopic level, (iv) Exploratory effort to examine shock induced phase changes in calcite single crystals. The single crystal calcite effort was motivated primarily by the comprehensive effort undertaken in Task (ii). Further discussion regarding these tasks is summarized below.

A. Nonlinear elastic response of shocked fused silica

Fused silica displays anomalous compression under both hydrostatic^{1,2} and shock loading³ conditions. The purpose of the present work was to use shear wave measurements to obtain a more complete description of the nonlinear elastic response of fused silica in the shocked state than is currently available.

Using in-material, particle velocity gauges in inclined, parallel plate impact experiments,^{4,5} large amplitude compression and shear wave profiles were measured in fused silica samples (Dynasil 1000). The longitudinal stress-strain relation calculated from the compression wave profiles was found to be identical over the stress range (to 60 kbar) of the present work to that reported for G.E. Type 151 fused silica³

Shear wave velocities, in the shocked state, were found to decrease linearly with compression up to a compression of 7.6 percent. At higher compressions, the shear wave velocities increased

with compression. These data in conjunction with the measured longitudinal stress-strain relation were used to obtain the longitudinal, shear and bulk moduli in the compressed state. An interesting finding was that the bulk and shear moduli display a minima at different density compressions. In addition to these moduli, the Poisson's ratio for shocked fused silica was determined as a function of density compression. It displays a complex, nonlinear behavior with values ranging between 0.17 and 0.06 over the 12 percent density compression range examined. This finding contradicts the assumption of constant Poisson's ratio for fused silica.⁶ and has interesting implications for stress deviators in shocked fused silica.

The mean stress versus density curve for shocked fused silica was calculated by integrating the bulk modulus versus compression curve. The mean stress-density compression curve under shock wave, uniaxial strain loading differs appreciably from the measured hydrostat and provides direct evidence for shear enhanced compaction in fused silica. This phenomenon is completely reversible to 60 kbar. The observed shear enhanced compaction appears to be compatible with a model proposed by Hemley et al.⁷ for the structural changes in hydrostatically compressed fused silica. Definitive support of this conjecture will require Raman measurements under shock loading.

Using the mean stress-density and longitudinal stress-density relation obtained in the present work, we obtain very large values of stress deviators in shocked fused silica. The large stress deviator values may play a significant role in the phase changes in fused silica observed at higher pressures.⁸

The present data were used to calculate second and third order elastic constants for fused silica. Two third order constants (C_{111} and C_{112}) were obtained from the present data. Two independent but consistent methodologies were used in analyzing our data. Both gave approximately the same results.

B. Shock induced phase transformation in polycrystalline calcite

Structural phase transformations under shock loading are of considerable interest for development of constitutive relations at high strain rates and for understanding the response of solids under nonhydrostatic stresses. Over the stress range where both phase changes and mechanical yielding can occur, it is difficult to separate these two phenomena from longitudinal wave profile measurements. Because crystal structures cannot be directly measured under shock loading, it is difficult to identify the phase transition mechanisms. Comparisons with static, high-pressure data are needed to interpret the shock data. These comparisons are difficult, for materials with significant strength, due to the difficulty in separating effects due to stress deviators and loading rates.

In the present work, the response of polycrystalline calcium carbonate was examined to assess the usefulness of measuring both shear and compression waves for better understanding shock-induced phase transformations in solids. In-material particle velocity histories were measured in Carrara marble subjected to combined compression and shear loading. These wave measurements presented the possibility of determining additional information on the shear response of the shocked material, and also the complete stress state produced by uniaxial strain loading. Both possibilities could contribute to improved understanding of shock-induced phase transformations by permitting more direct comparisons with static, high-pressure data, and providing greater information for development of material models. Specific issues were addressed concerning the dynamic response of polycrystalline calcium carbonate. This material was chosen for this initial application of the experimental method to a phase-transforming material because extensive static, high-pressure and shock wave experiments had been carried out previously,⁹⁻¹² and the available samples simplified execution and analysis of the experiments.

The experimental methods developed earlier^{4,5} for measuring in-material particle velocity histories under combined compression and shear loading were used with only a few modifications. A *new experimental development* was the implementation of a suggested technique¹³ for simultaneously measuring the two individual particle velocity components in a single experiment. This development permits precise time-correlation between the shear wave and the complex-structured compression wave that develops in a phase-transforming material.

Time-correlated shear and compression waves were measured in the marble at seven peak stress levels between 0.5 GPa and 5 GPa. The histories, measured at multiple depths within each sample, included the complete interval of compression and unloading. From these the shear, compression, and longitudinal release wave speeds were determined.

The shear waves were highly dispersive and were quenched by longitudinal unloading. The marble supported finite amplitude shear waves over the entire range investigated (to 11% density compression and 4.7 GPa longitudinal stress), which indicated that it retained significant strength. Above 3 GPa longitudinal stress, the shear wave amplitude decreased substantially with propagation distance.

The longitudinal waves had the complex structure observed in previous measurements,^{12,14} and were not self-similar. The compression wave first rises steeply, and then slowly, forming a knee. The continuous, gradual rise after the knee leads up to a second steep portion of the wave. The longitudinal release wave begins with a dispersive portion that leads down to a sharp rarefaction shock wave. The present measurements revealed a break in the rise of the compression wave for peak amplitudes below the knee. This break, and the knee in the higher-amplitude

experiments, attenuated with propagation distance. Hence, the compression waves were not self-similar.

A surface fitting technique was developed for calculating longitudinal stress and density in unsteady compression waves having a wide variety of shapes. These calculations indicated that lack of self-similarity in the wave had substantial effects on the stress and density determination below 1.55 GPa longitudinal stress. The locus of peak longitudinal stress-density states was found to approach the hydrostat for single crystal calcite for stresses above 2.5 GPa, as might occur if the same phases were produced under shock and hydrostatic loading.

The shear wave speeds in shocked Carrara marble as a function of density agreed well with the ultrasonic wave speed in hydrostatically compressed Oak Hall limestone. The decrease in the shear wave speed observed at small compressions substantiates earlier interpretations based on longitudinal wave measurements^{14,15} that the $\text{CaCO}_3(\text{II})$ phase is produced under impact loading. At higher compression, the shear modulus in the shocked marble increases with compression. This occurs after the marble has undergone a phase transformation. This finding contradicts the inferences in earlier work.¹⁴

The longitudinal wave speeds in shocked Carrara marble as a function of density were in good agreement with the speeds determined in Oak Hall limestone under shock¹² and hydrostatic loading⁹ to 9% density compression. At 11% compression, the longitudinal wave speed in the shocked marble was below an extrapolation from the hydrostatic data.

The shear modulus and bulk modulus in the shocked marble were determined from the measured wave speeds and calculated peak densities. The bulk modulus in the shocked marble as a function of density was in reasonable agreement with hydrostatic data up to 9% compression, which is near the onset on the $\text{CaCO}_3(\text{III})$ phase stability field under hydrostatic compression. Beyond this, at 11% compression, there is a pronounced discrepancy between the bulk modulus in the shocked marble and the bulk modulus determined from the hydrostatic compression curve for single crystal calcite. No such discrepancy was indicated by the trend of the peak stress-density states towards the hydrostat. However, the comparison of bulk modulus data from shock and static experiments is direct and unambiguous. The occurrence of a discrepancy in this comparison with no corresponding discrepancy in the comparison of the peak stress states with the hydrostat indicates that the latter comparison must be interpreted with considerable caution.

In contrast to previous studies,^{16,17} the mean stress-density relation for the shocked marble is not the integral of the bulk modulus-density relation. Failure to account for the systematic difference between the frozen phase-composition modulus, which was obtained from the wave speeds in

the shocked marble, and the relaxed modulus, which gives the slope of the mean stress-density relation, lead to an overestimate of the mean stress.

A model of the material's mean stress response was presented to illustrate a method for inferring the mean stress-density relation for shocked materials that undergo phase transformations from the experimentally determined bulk modulus-density relation. The model was a variation of the one developed by Grady et al.¹² to describe the response up through the $\text{CaCO}_3(\text{II})$ phase, taking the heterogeneity of the stress field in the polycrystal into account.

Based on the calculated density, the majority of the transformation occurs in the second steep rise in the compression wave, above a particle velocity of 115 m/s. The estimates of the longitudinal stress and density during unloading, suggested that the rarefaction shock is produced by direct reversion to calcite of either $\text{CaCO}_3(\text{II})$ or the succeeding high-pressure phase, depending on the peak state of compression.

Comparing measured shear wave amplitudes with previous wave propagation simulations¹⁸ suggested that the marble exhibits pressure-dependent yielding. This comparison indicated the usefulness of analyzing the shear wave amplitudes, as well as its complexity. The yield models used in the simulations were insufficient to quantify the marble strength or identify rate dependence in the yielding.

Based on a strong similarity between measured and simulated¹⁸ shear waves, it was argued that the compression wave, alone, causes yielding in the marble under combined compression-shear loading at a longitudinal stress below 2 GPa. The break in the compression wave that occurred below 70 m/s longitudinal particle velocity suggests that the compression wave, alone, may cause yielding at only 0.7 GPa longitudinal stress.

The present work has demonstrated that shear waves can be measured in a shock-induced, high-pressure phase, providing additional information not obtainable from uniaxial strain, plate impact experiments. The data determined from analyses of the measured waves facilitated new and more direct comparisons with static measurements. The difference in bulk modulus values beyond 9% compression cannot be explained by a difference between the dynamic mean stress at which $\text{CaCO}_3(\text{III})$ is produced and the equilibrium transformation pressure. It suggests the onset of a transformation to a phase having a much lower bulk modulus than does $\text{CaCO}_3(\text{III})$. Comparison of longitudinal stress-density states with a hydrostat is difficult to interpret, and was found to be misleading when judged against the direct comparison of bulk modulus data.

C. Time-resolved optical spectroscopy in shocked solids

The work on fused silica and polycrystalline calcite has clearly shown the value of shear wave measurements to characterize the shocked state. However, there exists a need for developing experimental methods to probe the shocked state at the microscopic level. Hence, an important objective of the present work was to explore the use of optical spectroscopic methods to obtain information at the atomic/molecular level. Because this is a new undertaking, the research effort was focused on developing experimental methods and performing feasibility experiments; we have emphasized time-resolved measurements in our work. The equipment procured under AFOSR funding has been necessary to undertake this activity.

In one set of experiments we developed an experimental method to obtain luminescence spectra during tension. Ruby samples were shocked along the crystal c-axis to create successive compression and tension loading using wave interactions. Time-resolved luminescence spectra were then obtained that corresponded to peak compression, complete unloading and peak tension states. In tension, the luminescence R lines show a blue shift and the line splitting increases with increasing tension. These data show that the sapphire lattice can sustain tensile stresses as high as 108 kbar (under uniaxial strain) without any damage. In addition, these data have been valuable to our theoretical effort to correlate optical spectra to symmetry changes at the local level. A comprehensive account of this theoretical effort will be published shortly.

In a second undertaking, we developed a method to do time-resolved, Raman measurements in shocked solids. Diamond was selected because of its large Raman cross-section¹⁹ and the considerable body of static high pressure measurements.²⁰⁻²² The frequency shift of the first order Raman line at $\omega_0 = 1333 \text{ cm}^{-1}$ was observed under uniaxial strain along the [110] direction to a longitudinal stress of 121 kbar. The measured frequency shift was consistent with the calculated strain-induced changes in the phonon frequency in diamond. Further improvements in spectral resolution are needed to observe the removal of the threefold degeneracy for compression along the [110] direction.

The present work on time-resolved, optical spectroscopy is a good start and makes a strong case for using this approach to better understand shock deformation in solids in future studies.

D. Phase changes in calcite single crystals shocked along the z-axis

This work was motivated by the results obtained for polycrystalline CaCO_3 summarized earlier. The objective of the present investigation was to undertake an exploratory effort, using single crystals, to examine the phase changes in calcite. Single crystal experiments avoid the difficulties due to stress concentrations arising from presence of randomly oriented grains and are necessary to

investigate detailed mechanisms. There have not been detailed studies of shocked calcite crystals over the compression range that are comparable to measurements under hydrostatic loading.^{10,11} Although it would be desirable to use particle velocity gauges, similar to the polycrystalline effort, available crystals did not allow for such measurements.

Quartz gauges^{23,24} were used to examine the shock response of calcite crystals compressed along the z-axis. Peak stress values ranged between 7 and 40 kbar. Stress time profiles were measured both at the impact surface and after transmission through 1-2 mm thick samples.

The measured longitudinal stress-particle velocity ($\sigma_x - u$) response for phase I is in good agreement with the calculated nonlinear elastic response using elastic constants cited in the literature.¹¹ These calculations also show the strong compressional anisotropy in calcite. The mean stress-density curve for uniaxial strain loading is below the hydrostatic loading curve. The calcite I→II transition is very rapid (few ns or less) and occurs at a longitudinal stress of approximately 17.5 ± 0.8 kbar. The density compression at the onset of transformation is 2.1 percent and is independent of the type of loading.

At the higher stresses (between 20-40 kbar), the shock response of calcite is complex and the transmitted profiles show a three wave structure with considerable rate dependence associated with the second transformation. The stress at which the second transformation occurs is considerably higher than the pressure for the calcite II→III transition under hydrostatic loading; the reasons for this difference are not understood. Because of the time-dependent features and the impedance differences between calcite and quartz, a detailed analysis of the transmitted profiles is not feasible. The impact surface measurements, in which the $\sigma - u$ data are measured at the calcite/quartz interface, also reveal time-dependent response beyond the calcite I→II transition. It is not possible to reconcile the impact surface and transmitted wave profile data with the stability fields for phases II and III under hydrostatic loading. Our data make a strong case for the occurrence of a final state that is different from the calcite III phase observed under hydrostatic loading. These findings support Aidun's results for the polycrystalline CaCO_3 .

Although our work has not been definitive over the 22-40 kbar range, it has provided interesting details and raised questions about present understanding regarding shock response of calcite. The present work suggests the need for alternate experiments for determining the nature of the shocked state. Because of the time-dependent response, comparison of the continuum data with static measurements is difficult and the need for spectroscopic results to get insight into the crystal structure under shock loading is emphasized.

REFERENCES

1. P.W. Bridgman, Am. J. Sci. 297, 7 (1939).
2. C. Meade and R. Jeanloz, Phys. Rev. 35, B236 (1987).
3. L.M. Barker and R.E. Hollenbach, J. Appl. Phys. 41, 4208 (1970).
4. Y.M. Gupta, Appl. Phys. Lett. 29, 694 (1976).
5. Y.M. Gupta, D.D. Keough, D.F. Walter, K.C. Dao, D. Henley and A. Urweider, Rev. Sci. Instrum. 51, 183 (1980).
6. H. Sugiura, K. Kondo and A. Sawaoka, "Anomalous compression mechanism of fused silica", in *High Pressure Research in Geophysics*, edited by S. Akimoto and M.H. Manghnani (Center for Publishing, Japan and D. Reidel Publishing, Co., 1982).
7. R.J. Hemley, H.K. Mao, P.M. Bell and B.O. Mysen, Phys. Rev. Lett. 57, 747 (1986).
8. J. Wackerle, J. Appl. Phys. 33, 922 (1962).
9. C.-Y. Wang, J. Geophys. Res. 71, 3543 (1966); C.-Y. Wang and M. Meltzer, J. Geophys. Res. 78, 1293 (1973).
10. A.K. Singh and G.C. Kennedy, J. Geophys. Res. 79, 2615 (1974).
11. D. Vo Thanh and A. Lacam, Phys. Earth Planet. Inter. 34, 195 (1984); D. Vo Thanh, Ph.D. thesis, Universite de Paris VII, 1981.
12. D.E. Grady, "Compression wave studies in Oakhall limestone", SAND83-0370, Sandia National Laboratories, Albuquerque, NM (March 1983).
13. J.W. Swegle, J. Appl. Phys. 49, 4280 (1978).
14. D.E. Grady, R.E. Hollenbach and K. Shuler, J. Geophys. Res. 83, 2839 (1978).
15. T.J. Ahrens and V.G. Gregson, J. Geophys. Res. 69, 4839 (1964).
16. M.P. Conner, M.S. thesis, Washington State University, 1988.
17. Y.M. Gupta, J. Appl. Phys. 51, 5352 (1980).
18. Y.M. Gupta, "Development of a method for determining dynamic shear properties", Draft Final Technical Report to the Defense Nuclear Agency under contract no. DNA 001-76-C-0384, 1978.
19. M. Cardona, "Light Scattering in Solids II", *Topics in Applied Physics V50*, edited by M. Cardona and G. Guntherodt (Springer-Verlag, New York, 1982), p. 93.

20. S.S. Mitra, O. Brafman, W.B. Daniels and R.K. Crawford, *Phys. Rev.* *186*, 942 (1969).
21. E. Whalley, A. Lavergne and P. Wong, *Rev. Sci. Instrum.* *47*, 845 (1976).
22. B.J. Parsons, *Proc. R. Soc. London. Ser. A* *352*, 397 (1977).
23. R.A. Graham, F.W. Nielson and W.B. Benedick, *J. Appl. Phys.* *36*, 1775 (1965).
24. Z.-P. Tang, Y.M. Gupta and P.M. Bellamy, *Rev. Sci. Instrum.* *59*, 1189 (1988).

III. SUMMARY OF THE LASER ENERGY DEPOSITION EFFORT

The investigation of non-linear laser pulse interaction with transparent solids culminated in explaining the fundamental mechanisms responsible for energy deposition and the material's response to it. As a result we have provided an explanation for single-shot and multi-pulse intrinsic laser damage in wide-gap high laser power optical materials.

In the course of this work new experimental techniques have been developed for the measurement of energy deposition from a laser pulse at visible and near infrared wavelengths into transparent solids. Their application to alkali halides and fused silica (SiO_2) revealed that lattice heating occurs via nonlinear absorption of laser photons by multiphoton excited free electrons with only small contributions that can be attributed to point defects created during the laser-solid interaction.

In contrast to these findings is our discovery that multi-pulse effects at intensities below the single-shot damage threshold are dominated by laser-generated stable defects. We have formulated a new model of intrinsic multi-pulse bulk damage that is consistent with our experimental observations. It is based on thermomechanical stress induced by accumulation of multiphoton generated F-centers.

The interaction of intense laser pulses with optical materials has been the subject of study for over two decades with the goal to explain the basic mechanisms of laser damage (optical dielectric breakdown) of intrinsically pure and transparent wide-gap solids. Alkali halides have traditionally been of particular interest in these investigations.

In discussing intrinsic optical dielectric breakdown in alkali halides one has to distinguish between single-pulse damage and catastrophic failure due to repetitive exposure of the same material to laser pulses of power density below the single pulse damage threshold. In both cases breakdown is the result of energy deposition from the laser beam to the solid associated with a sequence of complex electronic and atomic (photo-chemical) processes, but, as we will show in this report, with significantly different importance of laser-induced defects.

The primary process in both cases is electron-hole pairs (exciton) formation by multiphoton interband transitions.¹ Single pulse damage is due to non-linear absorption by the resulting free electrons with only small contributions from absorption by primary point defects that are produced as follow-on products of exciton recombination. In contrast to this situation, the shot to shot accumulation of stable point defects and the associated thermo-mechanical stresses turned out to be the cause of multi-pulse failure.¹

Historically, the mechanism of intrinsic laser damage was thought to be avalanche impact ionization by free electrons which had gained sufficient kinetic energy during the interaction with the

intense laser photon field.² The origin of this concept can be traced to the work by von Hippel³ and others who believed dc dielectric breakdown occurs by this process. Forty years later, Yablonovitch and Bloembergen noted an apparent agreement of laser damage field strengths (all smaller than 20 MV/cm) at 10.6 μm and dc data. This led them to propose an early version of the so-called electron avalanche laser breakdown theory⁴ which until very recently dominated the thinking of virtually every worker in the field.⁵

The picture of dielectric breakdown at dc to optical frequencies has dramatically changed in the last five years due to the work of Fischetti, DiMaria, Cartier, and co-workers at IBM and our group at Washington State University. Cartier et al.⁶ were able to measure the electron-photon scattering rates in amorphous SiO_2 as a function of electron energy, confirming calculations by Fischetti et al. and enabling Monte Carlo simulations at high dc field of electron transport in a number of wide-gap solids.⁷ DiMaria and co-workers developed methods for the direct measurement of electron energy distributions under high field conditions.⁸ In agreement with Fischetti's predictions they found that the energy distributions provided *no evidence for electron multiplication (avalanche formation)* up to dc field strengths of 20 MV/cm.⁹ It turned out that efficient scattering of electrons above 2 eV with acoustic phonons stabilizes the electron energy distribution and prevents runaway.¹⁰ Similar conclusions are expected to hold for alkali halides.

Independent of the fundamental investigations at IBM we reached similar conclusions concerning intrinsic damage in NaCl, KBr, KI, and SiO_2 at optical frequencies. We were able to develop experimental techniques for the *measurement of prebreakdown energy deposition from the laser pulse into intrinsically transparent solids*. These made possible the systematic investigation of the processes involved in the deposition of energy from the laser beam into the transparent solid (including the role of laser-generated primary defects) and provided hard evidence for the absence of avalanche formation at visible and near-infrared wavelengths up to the intrinsic single-shot damage threshold.

A. The mechanism of single pulse laser damage in alkali halides

Two types of experiments were performed in the prebreakdown regime to measure the absorption of laser beam energy, and these measurements were compared with models including processes that are known to occur in alkali halides following electron-hole pair production. Two alternative models for free-electron heating were investigated. The difference in (theoretical) heating efficiency of these two lead to conclusions regarding the relative importance of primary defects in single-shot laser damage.

Calibrated photoacoustic measurements of prebreakdown energy absorption from 532 nm laser pulses in NaCl revealed that the primary process for the interaction is four-photon absorption. These results are compared with three models of the interaction.

They account in detail for the creation and recombination of e-h pairs, creation and annihilation of primary defects (V_k , STE, STH, F), and the resulting heating of the lattice. We allow for two possible pair generation processes, multiphoton absorption and impact ionization. Generally, electrons are elevated to the conduction band either from the valence band or from ionized defects (STE's, F-centers). Free valence band holes are trapped rapidly, but may be freed again by hole absorption, then quickly retrapped. The net result is heat deposition of $\hbar\omega$ per event. V_k centers may orient themselves into directions relatively inert to the polarized laser light, thus the hole absorption does not occur for each V_k center throughout the duration of the laser pulse.

F-center formation is assumed to occur through the triplet STE state at high temperatures. This effect is included or excluded as to its interest in the particular experiment modeled.

The two treatments of free-carrier heating are called (1) the free-electron model, a nonlinear absorption of electromagnetic energy by fairly energetic conduction electrons and (2) the polaron model, wherein low energy conduction electrons which are *strongly coupled to lattice ions* absorb photons linearly. The first is vastly more efficient theoretically, but there is no *a priori* reason to prefer one of these unproven models. For completeness, we also compared the data with a model including electron-impact ionization avalanche production of e-h pairs.

Both treatments of free carrier heating satisfactorily represent the measurements. The four-photon absorption cross-section is ambiguous because the photoacoustic effect is a result of total absorbed energy. Allowing for secondary absorption processes (in addition to four-photon absorption) without a preference for one of the two alternatives, it cannot be determined which value of $\sigma^{(4)}$ is most correct, since both are satisfactory. If model (1) is correct, then fewer electrons, and therefore, fewer primary defects are required to produce the effect. In fact, for model (1), $\approx 92\%$ of all energy absorption is via free electron heating. In model (2) defect absorption, primarily V_k center absorption, accounts for most of the heating ($> 50\%$). Further, the avalanche process does not account for the data.

In order to discover which model is most correct, and thus the importance of primary defects, it is necessary to model similar measurements with a known value of $\sigma^{(4)}$. This was accomplished using the self-trapped exciton recombination luminescence or STERL technique.

The STERL technique of measuring multiphoton excitation takes advantage of this intrinsic radiative relaxation of electron-hole (e-h) pairs in alkali halides. Since the process is intrinsic, the total light signal output is a measure of the number of e-h pairs generated in an excitation event.

This then represents an improvement in measuring multiphoton cross-sections over the photoacoustic method because, in a limited range of incident pulse intensities, the response of the sample to incident ionizing radiation is proportional only to the total number of excited carriers. This proportionality falls off when the crystal temperature exceeds that at which non-radiative recombination processes become important. This effect was used as a thermometer to determine lattice heating in KBr when pulse energies were increased beyond the level where thermal quenching of the luminescence begins. Armed with the value of $\sigma^{(4)}$ obtained at low pulse intensities, the multiphoton-polaron and -free electron theories of energy absorption were compared with experimental data, revealing that the polaron-defect absorption is simply too weak to account for the observed temperature increase, leaving the multiphoton-free-carrier model as a complete, satisfactory model of prebreakdown energy absorption. Further, temperature data indicate that the highest intensity *nondamaging* laser pulses induced heating in KBr to only a small amount below the melting point. These measurements could all be explained and modeled without any necessity of including avalanche generation. In fact, these measurements and calculations preclude the role of avalanche generation at least up to the melting point, even though electron densities greater than 10^{18} cm^{-3} are present. These results were verified in KI (three photon band gap at 532 nm).

The conclusion reached in the STERL study was bolstered by a third experiment. Pulses of 266 nm laser light were used to create e-h pairs in NaCl and SiO₂ (via 2-photon absorption), and the photoacoustic signal was monitored as a function of coincident pulse energies of 1064 nm laser light. Since this wavelength is too long to create e-h pairs, the only mechanism by which absorption could take place is via conduction carriers. The nonlinear nature of the resultant signal verified the correctness of the free-electron treatment of the conduction carrier absorption, thus relegating the participation of defects to a minor or possible insignificant role in the single hot damage process. Exaggerating their roles in the model calculations showed that the models would cease to represent the data. Therefore, we conclude primary defects play at most a minor role in single pulse laser damage of pure alkali halides.

The work presented up to this point reflects significant advances in the understanding of the fundamental interactions between high power pulsed laser beams and transparent solids. Identifying the intrinsic single shot damage threshold provides an important standard of performance for optical materials. But perhaps of greater importance from a practical standpoint are the processes contributing to multi-pulse laser damage. In the next section, we explore a new model of multiple-pulse-on-one-site damage based on the accumulation of laser induced color center formation. This new model, an out-growth of the model calculations discussed in previous sections, is not concerned with electrons but in the alteration of the mechanical properties of the crystalline solid which accompany the formation of lattice defects.

B. Multiple shot bulk damage

In contrast to single shot experiments, where temperature increases of several hundred degrees were observed,¹ during the multiple pulse experiments presented here the peak temperature increase is at most a few tens of degrees. Under these conditions it is clear that a different failure mode is required to explain the eventual damage of a crystal at laser power levels well below the single shot damage threshold. The most obvious cumulative effect which might suffice to cause damage at power levels below the single shot damage threshold is the buildup of F-centers as a result of laser-induced electron-hole pair generation.

In alkali halides, F-centers have a typical absorption peak of 2-3 eV and can, therefore, release their electrons into the conduction band through single or two-photon absorption at visible frequencies. However, Jones et al.¹ have shown that in alkali halides the effect of F-centers on lattice heating is insignificant, even with relatively large initial concentrations. Thus, at first, it would appear that F-center accumulation is not responsible for the observed multi-shot damage behavior in KBr and KI reported here. However, we now suggest an alternative mechanism in which these defects can make major contribution in the damage process.

It has been shown in alkali halides that when a crystal is radiatively colored it undergoes an expansion due to the production of Frenkel defects. As F-centers accumulate in the focal volume from shot to shot, local stresses will develop since the associated expansion is impeded by the surrounding uncolored crystal. If this stress reaches a sufficient level it is expected that the material will crack or experience plastic flow resulting in a nonuniformity in the crystal which acts as a scattering center. Scattering of the incident laser pulse from this nonuniformity is detected and interpreted as damage.

The model presented here assumes that failure occurs when the local stress, $\bar{\sigma}$, exceeds the yield stress, σ_y . To simplify matters we take the local stress to be the mean of the diagonal elements of the stress tensor, or $\bar{\sigma} \equiv (\sigma_{11} + \sigma_{22} + \sigma_{33})/3$. The yield stress, however, is not independent of defect concentration. Alkali halide crystals exhibit an increase in the yield stress after radiative coloring. Nadeau et al.¹¹ found the increase in yield stress to be proportional to the square root of the F-center concentration, n_F , or

$$\sigma_y = A + B\sqrt{n_F}, \quad (1)$$

with $A = 6.47 \times 10^6$ dyne cm^{-2} and $B = 6.54 \times 10^{-2}$ dyne $\text{cm}^{-1/2}$ for KBr.

The induced stress consists of two components of different origin. The first is due to the lattice expansion in the focal volume caused by the production of F-centers. The associated volume increase, v , due to the production of one F-center has been determined by measuring the linear

expansion of a crystal as a function of F-center concentration: $v = 0.074a^3$ for NaCl and KBr (a^3 is the volume of the original, undisturbed lattice cell).

In calculating the thermal stress, σ_T , we are able to make use of our single shot results which showed that the peak temperature increase, during the laser pulse, is very nearly proportional to F_0^m , where F_0 is the peak photon flux density, and m is the order of multiphoton absorption. Therefore, the thermal stress is

$$\bar{\sigma}_T \approx C\alpha KF_0^m. \quad (2)$$

Here, α is the coefficient of thermal expansion, C is a constant of proportionality and F_0 is the peak photon flux. The total stress is the sum of these two components

$$\bar{\sigma} = \bar{\sigma}_F + \bar{\sigma}_T. \quad (3)$$

It remains to determine the growth of the F-center concentration. Drawing an analogy to the failure of materials under load Zhurkov et al.¹² and Manenkov et al.¹³ have suggested that multi-shot damage may be due to a mechanochemical reaction governed by a reaction rate of the form, $dN/dt \approx k_b T \exp\{-(U_0 - \gamma\sigma)/k_B T\}$. N represents the population of the appropriate reaction product associated with the damage process, k_B is Boltzmann's constant, T is the temperature, and U_0 is the activation energy of the process. σ is then the applied stress and γ is a material dependent parameter. The effect of the stress is to reduce the activation energy for the nucleation process. We propose that F-center production is the pertinent mechanochemical process to which the above rate applies. Indeed, there have been studies where an increase in F-center production with applied stress was observed.

The number of F-centers produced during any one laser pulse under stress free conditions is a known function of temperature. F-center production is also proportional to the number of electron-hole pairs produced. For KBr and KI at 532 nm, this is in turn proportional to F_0^m . Thus, for any one laser pulse

$$\Delta n_F = DF_0^m k_B T \exp\left\{-\left[U_0 - \gamma\bar{\sigma}\right]/k_B T\right\}, \quad (4)$$

with D being a constant of proportionality, which is determined from the known, stress free, defect production efficiencies. An iterative calculation is then used to follow the buildup of F-centers and stress from one shot to the next until the condition for failure is met.

The experimental procedure is to expose the sample to a series of laser pulses of uniform peak power at a repetition frequency of about one Hertz. The laser pulses are Gaussian in both spatial and temporal profiles, with a spot size of 8-10 μm and a pulse length of approximately 70 psec.

These values refer to the diameter and half-width at $1/e$ of the peak photon flux density, respectively. The peak flux density varied from about 0.9×10^{29} to 1.6×10^{29} photons $\text{cm}^{-2} \text{sec}^{-1}$ (34-52 GW/ cm^2) for the experiments on KBr and ranged from 0.6×10^{29} to 1.0×10^{29} photons $\text{cm}^{-2} \text{sec}^{-1}$ (22-37 GM/ cm^2) for KI. The sample is mounted on the cold finger of a closed cycle, optical cryostat and the temperature can be varied from about 50 K to room temperature. The condition of the crystal is monitored by measuring the scattering of laser light perpendicular to the beam axis with a photomultiplier tube. Damage is assumed to have occurred when a sudden increase in light scattering is observed. The decay time for the thermal disturbance is on the order of 10^{-5} sec for the focal spot size used in the experiment, thus precluding a simple buildup of temperature at the pulse repetition frequency used.

It is important to differentiate between data which represent the intrinsic behavior of the material and those which are extrinsic. The intrinsic behavior represents a fundamental limit of the crystal, whereas the extrinsic behavior can be caused by any number of defects or impurities initially present in the crystal. Although for any give photon flux density there may be a large variation in the number of shots withstood by different sites in the sample, all data points are seen to lie within a distinct envelope. Rather than performing a statistical analysis of the variations we feel that, given a sufficient number of points, the intrinsic behavior of the crystal becomes evident. The intrinsic limit exhibits itself as a boundary beyond which no data points fall.

This procedure, however, can only be applied to crystals of very high purity. Otherwise, there may be too few sites which exhibit intrinsic behavior for the envelope of points to become clear. For this reason we use crystals from the University of Utah grown with reactive atmosphere processes, ultra pure material.

At low initial temperatures, where the efficiency of F-center production is very small, the thermal stress is particularly important. In this case the stress must reach a sufficient level to boost the F-center formation efficiency to start the accumulation process, and we see a distinct threshold-like behavior. At higher temperatures the F-center formation rate is large enough (without being enhanced by the thermal stress) so that there is a much weaker flux dependence.

Results of single shot experiments indicate that defects play, at most, a minor role in the heating (and thus damage) of wide bandgap, optical materials. However, at the power levels used in multishot experiments, heating alone is not sufficient to cause damage. In this case the accumulation of defects from shot-to-shot leads to a buildup of stress and, ultimately, the failure of the material. We have developed a model for the behavior of KBr and KI exposed to multiple laser pulses induced in the crystal. The model applies to the intrinsic behavior of the crystal in that it does not appeal to a probabilistic mechanism or to the inclusion of absorbing impurities or defects.

Instead, it attempts to incorporate known responses of the crystal to the laser pulses, temperature and stress. At this stage in its development this model predicts the correct dependence of multi-shot laser damage on temperature and photon flux density for the two materials studied. However, additional refinements to the model (such as; the inclusion of strain rate effects and the possibility of defect clustering) and experimental confirmation of the defect accumulation as described in the model are required before a final assessment of its validity can be made.

REFERENCES

1. S.C. Jones, P. Braunlich, R.T. Casper, X.-A. Shen, and P. Kelly, *Opt. Eng.* **28**, 1039 (1989).
2. N. Bloembergen, *IEEE J. Quantum Electron.*, QE-10, 375 (1974).
3. A. von Hippel, *Z. Phys.* **75**, 145 (1932).
4. A. Yablonovitch and N. Bloembergen, *Phys. Rev. Lett.* **29**, 907 (1972).
5. W.L. Smith, *Opt. Eng.* **17**, 489 (1978).
6. E. Cartier and P. Pfluger, *Physica Script T23*, 1988 (Trends in Physics EPS-7); and J., Bernastoni, E. Cartier and P. Pfluger, *Phys. Rev.* **B38**, 12567 (1988).
7. M.V. Fischetti, *Phys. Rev. Lett.* **53**, 1755 (1984); and M.V. Fischetti, D.J. DiMaria, S.D. Brorson, T.N. Theis, and J.R. Kirtley, *Phys. Rev.* **B31**, 8124 (1985).
8. D.J. DiMaria and M.V. Fischetti in *The Physics and Chemistry of SiO₂ and the Si-SiO₂ Interface*, edited by C.R. Helms and B.E. Deal (Plenum Press, New York, 1988).
9. S.D. Brorson, D.J. DiMaria, M.V. Fischetti, F.L. Pesavento, P.M. Solomon, and D.W. Dong, *J. Appl. Phys.* **57**, 1302 (1985).
10. D.J. DiMaria, M.V. Fischetti, and E. Tierney, *Phys. Rev. Lett.* **56**, 1284 (1986).
11. John S. Nadeau, *J. Appl. Phys.* **35**, 1248 (1964).
12. S.N. Zhurkov, S.B. Eron'ko, and A. Chmel', *Fiz. Tverd. Tela 3040 (1980) [Sov. Phys. Solid State 22, 1776 (1980)]*.
13. A.A. Manenkov, G.A. Matyushun, V.S. Nechtailo, and A.S. Tsaprilov, *Kvantovaya Elektron*, **10**, 2426 (1983) [*Sov. J. Quantum Electron.* **10**, 1580 (1983)].

IV. CONCLUDING REMARKS

The shock wave effort has demonstrated the usefulness of making shear wave measurements for understanding the nonlinear response of materials under dynamic loading. In fused silica, the shear wave measurements led to the finding of reversible, shear-enhanced compaction. In addition, stress deviators determined from our work are considerably different from those assumed in present studies.

The work on polycrystalline calcite represents the first study to use shear wave measurements to understand shock induced phase changes. The shear wave measurements have provided new information about the calcite response that contradicts existing inferences about the shock response in calcite. The direct measurement of shear modulus decrease, associated with the calcite I→II transition, is an important result that confirms the occurrence of this transition. More importantly, the subsequent increase in shear modulus at higher compressions was a surprising result. This measurement underscores the need to quantify the shocked state in more detail and to use caution in comparing the $\sigma_x - V$ results under shock loading with static P - V data to understand the shock response. The possibility that a phase other than calcite III is being formed under shock loading is an interesting one and that needs to be pursued in future studies. More definitive conclusions will require optical spectroscopy measurements in conjunction with use of calcite single crystals. The small effort undertaken on the shock measurements in calcite single crystals reveals clearly the I→II transition. However, the response between 22-40 kbar is complex and characterized by time dependent transformation to at least one or more phases. Continuum measurements alone are insufficient to resolve this response.

During the course of the present work, we demonstrated the ability to do time-resolved, optical spectroscopy experiments in shocked solids. Both luminescence and Raman measurements can now be carried out. These developments represent new approaches to characterizing the shocked state and are complementary to the continuum measurements. The combination of the two types of measurements will be required to better understand the shocked state.

The laser effort was successful in clearing up several confused issues in the field of interaction of intense laser pulses with intrinsically transparent optical materials. In fact, the physical processes responsible for laser damage due to energy deposition from the laser pulse into the transparent solid were explained by carefully executed measurements in the prebreakdown power density regime. As a result, we have shown that the presumption of laser damage due to laser-induced electron avalanche formation is not needed to explain intrinsic, single pulse bulk damage for $E_g/\hbar\omega < 5$ (E_g is the width of the forbidden gap of the solid) and that avalanches are not measurably formed in NaCl and SiO₂ for $E_g/\hbar\omega \geq 7$ even when large concentrations of free electrons are

present and the rms electric field is near the breakdown value. Instead, the process at visible wavelengths is satisfactorily explained by laser energy absorption by multi-photon-excited free electrons. Damage correlates with lattice heating to near the melting point.

Based on our results, we can define the intrinsic, single-shot, bulk damage for three alkali halides at $\lambda = 532$ nm, $\tau = 80$ –100 psec

$$\text{KI} : \sigma^{(3)} = (6 \pm 5) \times 10^{-81} \text{cm}^6 \text{sec}^2 \quad I_B = 75 \pm 7.5 \text{ GW/cm}^2$$

$$\text{KBr} : \sigma^{(4)} = (2 \pm 1) \times 10^{-112} \text{cm}^8 \text{sec}^3 \quad I_B = 240 \pm 24 \text{ GW/cm}^2$$

$$\text{NaCl} : \sigma^{(4)} = 1.5 \times 10^{-114} \text{cm}^8 \text{sec}^3 \quad I_B = 745 \pm 75 \text{ GW/cm}^2$$

The reported errors are simple uncertainties in pulse energy, beam radius and pulse length.

Within the multiphoton initiated damage model, dilute impurities or defects with interband electronic states do not make a significant contribution to single pulse damage in terms of energy absorption or providing free electrons. However, laser-induced lattice defect formation at intensities below the single shot damage threshold appears to be a plausible explanation of *multipulse* optical damage. This new avenue of investigation leads away from the idea that defects provide avalanche initiating electrons, thus, simply lowering the *single shot threshold*.

Using the knowledge of single pulse interactions as a starting point, we have developed an explanation for multishot bulk damage based on the response of the crystal to the laser induced temperature distribution and the accumulation of stable defects. The experimental techniques we have developed have been met with great interest by the so-called laser damage community which convenes annually at Boulder, CO for the Laser Damage Symposium. They were transferred to the Advanced Drivers Program of the Internal Confinement Fusion Effort and the Y-Division (Lasers) at the Lawrence Livermore National Laboratory (Contacts: Drs. Mark R. Kozlowski and C. Robert Wolfe). The intended applications there are in the study of laser pulse interaction with dielectric coatings on high power laser optical components.

Our investigations of the response of transparent optical materials to rapid energy deposition from intense laser pulses created a firm experimental basis for the understanding of the fundamental processes involved in dielectric breakdown at optical frequencies. This understanding led away from the traditional picture of avalanche breakdown, and the agreement of our work with recent new conclusions concerning dc breakdown is very gratifying.

We conclude by noting that understanding the dynamics of the laser damage interaction in multiple shot experiments will require time-resolved optical measurements to quantify the thermo-mechanical stresses around the defects. The time-resolved, optical spectroscopy measurements under the well defined conditions of plane shock wave loading are a start in this direction.

APPENDIX A

Publications and Presentations

Theses

1. Xiao-An Shen, "Intrinsic optical breakdown in KBr at 532 nm", Ph.D. dissertation, Washington State University (1987).
2. Mark P. Conner, "Shear wave measurements to determine the nonlinear elastic response of fused silica under shock loading", M.S. thesis, Washington State University (1988).
3. Lin Simpson, "Measurement of the three photon absorption cross section and intrinsic optical breakdown for KI at 532 nm", non-thesis M.S. report, Washington State University (1988).
4. John B. Aidun, "Study of shear and compression waves in shocked calcium carbonate", Ph.D. dissertation, Washington State University (1989).
5. Richard Thomas Casper, "Intrinsic multiple-shot laser-induced bulk damage in alkali halides at 532 nm", Ph.D. dissertation, Washington State University (1990).
6. Brad S. Stapleton, "Response of single crystal calcite shocked to 40 kbar along the z-axis", M.S. thesis, Washington State University (1990).

Publications

1. X.A. Shen, Scott C. Jones, Peter Braunlich and Paul Kelly, "Four-photon absorption cross section in potassium bromide at 532 nm", *Phys. Rev. B*: **36**, 2831 (1987).
2. X.A. Shen, Peter Braunlich, Scott C. Jones and Paul Kelly, "Intrinsic optical damage in KBr at 532 nm", *Phys. Rev. Lett.* **59**, 1605 (1987).
3. X.A. Shen, Peter Braunlich, Scott C. Jones and Paul Kelly, "Investigation of intrinsic optical damage in potassium bromide at 532 nm", *Phys. Rev. B*: **38**, 3494 (1988).
4. Peter Braunlich, Scott C. Jones, X.A. Shen, R. Thomas Casper and Paul Kelly, "The discovery of laser-induced intrinsic optical damage in wide-gap materials at visible wavelength", in *Laser Induced Damage in Optical Materials: 1987*, Nat. Inst. Stand. Tech. Spec. Publ. No. 756, edited by H.E. Bennett, A.H. Guenther, D. Milam, B.E. Newnam, M.J. Soileau (U.S. GPO, Washington, DC, 1988) p. 476.

5. R. Thomas Casper, Scott C. Jones, X.A. Shen, Peter Braunlich and Peter Kelly, "The laser damage mechanism for NaCl and KBr at 532 nm", in *Laser Induced Damage in Optical Materials: 1987*, Nat. Inst. Stand. Tech. Spec. Publ. No. 756, edited by H.E. Bennett, A.H. Guenther, D. Milam, B.E. Newnam, M.J. Soileau (U.S. GPO, Washington, DC, 1988) p. 485.
6. X.A. Shen, Peter Braunlich, Scott C. Jones, and Paul Kelly, "Intrinsic optical damage in potassium bromide at 532 nm", in *Laser Induced Damage in Optical Materials: 1987*, Nat. Inst. Stand. Tech. Spec. Publ. No. 756, edited by H.E. Bennett, A.H. Guenther, D. Milam, B.E. Newnam, M.J. Soileau (U.S. GPO, Washington, DC, 1988) p. 465.
7. John B. Aidun and Y.M. Gupta, "Shear-wave measurements for improved characterization of shock-induced phase transformations in Carrara marble", *Geophys. Res. Lett.* **16** (2), 191 (1989).
8. John B. Aidun and Y.M. Gupta, "Simultaneous measurement of in-material longitudinal and transverse particle velocity histories in a compression-shear experiment", *J. Appl. Phys.* **65** (5), 1898 (1989).
9. Y.M. Gupta, P.D. Horn and C.S. Yoo, "Time-resolved Raman spectrum of shock-compressed diamond", *Appl. Phys. Lett.* **55**, 33 (1989).
10. Scott C. Jones, Peter Braunlich, R. Thomas Casper, Xiao-An Shen and Paul Kelly, "Recent progress on laser-induced modifications and intrinsic bulk damage of wide-gap optical materials", *Opt. Eng.* **38**, 1039 (1989).
11. R. Thomas Casper, Scott C. Jones and Peter Braunlich, "Multiple shot intrinsic bulk damage in KBr at 532 nm", in *Laser Induced Damage in Optical Materials: 1988*, Nat. Inst. Stand. Tech. Spec. Publ. No. 775, edited by H.E. Bennett, A.H. Guenther, B.E. Newman and M.J. Soileau (U.S. GPO, Washington, DC, 1989) p. 12.
12. X.A. Shen, Scott C. Jones and Peter Braunlich, "Laser heating at free electrons and the mechanism of intrinsic laser breakdown in wide-gap optical materials at 1064 nm", in *Laser Induced Damage in Optical Materials: 1988*, Nat. Inst. Stand. Tech. Spec. Publ. No. 775, edited by H.E. Bennett, A.H. Guenther, B.E. Newman and M.J. Soileau (U.S. GPO, Washington, DC, 1989) p. 22.
13. Lin Simpson, X.A. Shen, Scott C. Jones, Peter Braunlich and Paul Kelly, "Measurement of the three photon absorption cross section and intrinsic optical breakdown of KI at 532 nm", in *Laser Induced Damage in Optical Materials: 1988*, Nat. Inst. Stand. Tech. Spec. Publ. No. 775, edited by H.E. Bennett, A.H. Guenther, B.E. Newman and M.J. Soileau (U.S. GPO, Washington, DC, 1989) p. 118.

14. X.A. Shen, Scott C. Jones and Peter Braunlich, "Laser heating of free electrons in wide-gap optical materials at 1064 nm", *Phys. Rev. Lett.* **62**, 2711 (1989).
15. Peter Braunlich, Scott C. Jones, Xiao-An Shen, R. Thomas Casper and Paul Kelly, "Laser-induced modifications and the mechanism of intrinsic damage in wide-gap optical materials", in *Nuclear Instruments and Methods in Physics Research B46*, (North Holland, 1990) p. 224.
16. R. Thomas Casper, Scott C. Jones, Peter Braunlich and Paul Kelly, "F-center accumulation as a mechanism of multiple-pulse, laser-induced bulk damage in KBr and KI at 532 nm", in *Nuclear Instruments and Methods in Physics Research B46* (North Holland, 1990) p. 231.
17. Peter Braunlich, Scott C. Jones, X.A. Shen, R. Thomas Casper and Paul Kelly, "The role of laser-induced defects in intrinsic laser damage", to be published in the *Proceedings of the Joint U.S.-Japan Seminar on Atomic Processes Induced by Electronic Excitations in Non-Metallic Solids*, Nagoya, Japan (September 1989).
18. P. Braunlich, S.C. Jones, X.A. Shen, R.T. Casper, D.J. DiMaria, M.V. Fischetti, E. Cartier and Paul Kelly, "Non-avalanche dielectric breakdown at DC and optical frequencies", in production for publication in *Proceedings of the 21st Annual Symposium on Optical Materials for High Power Lasers*, Boulder, CO (November 1989).
19. X.A. Shen and Y.M. Gupta, "Time-resolved luminescence measurements in ruby shocked along the crystal A-axis", to be published in *Shock Waves in Condensed Matter — 1989*, edited by S.C. Schmidt, J.N. Johnson and L.W. Davison (Plenum, New York, 1990).
20. M.P. Conner and Y.M. Gupta, "Shear wave measurements to determine the nonlinear elastic response of shocked fused silica", to be submitted to *J. Appl. Phys.*
21. J.B. Aidun and Y.M. Gupta, "Lagrangian analysis of particle velocity profiles in shocked calcite", to be submitted to *J. Appl. Phys.*
22. J.B. Aidun and Y.M. Gupta, "Shear and compression wave measurements to characterize shock induced phase transformations in Carrara marble", to be submitted to *J. Geophys. Res.*

Presentations

1. Y.M. Gupta, "Stress measurements in shock wave experiments: use of piezoresistance gauges", Invited Paper at the 1987 ASME Applied Mechanics, Bioengineering, and Fluids Engineering Conference, Cincinnati, OH (June 1987).

2. Y.M. Gupta, "Recent developments in dynamic stress measurements: an overview and some thoughts", Invited Presentation at the DNA INWET Conference, Arlington, VA (October 1987).
3. Peter Braunlich, Scott C. Jones, X.A. Shen, R. Thomas Casper and Paul Kelly, "The discovery of laser-induced intrinsic optical damage in wide-gap materials at visible wavelength", presented at the 19th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1987).
4. R. Thomas Casper, Scott C. Jones, X.A. Shen, Peter Braunlich and Paul Kelly, "The laser damage mechanism for NaCl and KBr at 532 nm - theoretical predictions and experimental tests", presented at the 19th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1987).
5. X.A. Shen, Peter Braunlich, Scott C. Jones and Paul Kelly, "Intrinsic optical damage in potassium bromide at 532 nm", presented at the 19th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1987).
6. J.B. Aidun, "Investigation of the effect of phase transition and inelastic deformation on the dynamic response of calcite", poster presented at the Symposium on Shock Wave Compression of Condensed Matter, Pullman, WA (September 1988).
7. J.B. Aidun and Y.M. Gupta, "Shear wave measurements in shock-induced high-pressure phases in polycrystalline CaCO_3 ", presented at the 1988 AGU Fall Meeting, San Francisco, CA (December 1988).
8. R. Thomas Casper, Scott C. Jones and Peter Braunlich, "Multiple shot intrinsic bulk damage in KBr at 532 nm", presented at the 20th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1988).
9. X.A. Shen, Scott C. Jones and Peter Braunlich, "Laser heating and free electrons and the mechanism of intrinsic laser breakdown in wide-gap optical materials at 1064 nm", presented at the 20th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1988).
10. Lin Simpson, X.A. Shen, Scott C. Jones, Peter Braunlich and Paul Kelly, "Measurement of the three photon absorption cross section and intrinsic optical breakdown of KI at 532 nm", presented at the 20th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (October 1988).

11. Y.M. Gupta, "Time-resolved optical and continuum measurements to probe the shocked state in condensed materials", presented at the University of Washington, Seattle, WA (May 1989).
12. John Aidun, "Shear wave measurements to characterize the shock induced phase transformations in polycrystalline calcite", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
13. P.F. Braunlich, "Historical review of laser damage studies: Theories, methods, controversies", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
14. R.T. Casper, "Models and measurements for single- and multi-pulse laser damage in alkali halides", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
15. M.P. Conner and Y.M. Gupta, "Shear wave measurements to determine the nonlinear elastic response of fused silica under shock loading", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
16. Y.M. Gupta, "Shock wave measurements in geophysical and ceramic materials: Limitations and needs", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
17. S.C. Jones, "Models for and measurements of multiphoton initiated prebreakdown energy absorption in wide gap insulators", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
18. X.A. Shen, "Multiphoton absorption, lattice heating and laser damage in alkali halides", presented at the AFOSR Workshop on Nonlinear Material Response to Very Rapid Energy Deposition, Pullman, WA (May 1989).
19. Peter Braunlich, Scott C. Jones, Xiao-An Shen, R. Thomas Casper and Paul Kelly, "Laser-induced modifications and the mechanism of intrinsic damage in wide-gap optical materials", presented at the Conference on Radiation Effects in Insulator - 5 (REI-5), Hamilton, Canada (June 1989).
20. R. Thomas Casper, Scott C. Jones, Peter Braunlich and Paul Kelly, "F-center accumulation as a mechanism of multiple-pulse, laser-induced bulk damage in KBr and KI at 532 nm", presented at the Conference on Radiation Effects in Insulators - 5 (REI-5), Hamilton, Canada (June 1989).

21. J.B. Aidun and Y.M. Gupta, "Inferring mean stress in phase-transforming, shocked material from measured shear and compression waves", presented at the APS Topical Conference on Shock Compression of Condensed Matter, Albuquerque, NM (August 1989).
22. M.P. Conner and Y.M. Gupta, "Shear wave measurements to determine the nonlinear elastic response of shocked fused silica", presented at the APS Topical Conference on Shock Compression of Condensed Matter, Albuquerque, NM (August 1989).
23. X.A. Shen and Y.M. Gupta, "Time-resolved luminescence measurements in ruby shocked along the crystal A-axis", presented at the APS Topical Conference on Shock Compression of Condensed Matter, Albuquerque, NM (August 1989).
24. J.B. Aidun, "Shear and compression wave measurements during structural phase transformations in calcite carbonate", presented at Battelle Pacific Northwest Laboratories, Richland, WA (October 1989).
25. P. Braunlich, S.C. Jones, X.A. Shen, R.T. Casper, D.J. DiMaria, M.V. Fischetti, E. Cartier and Paul Kelly, "Non avalanche dielectric breakdown at DC and optical frequencies", presented at the 21st Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO (November 1989).
26. Y.M. Gupta and M.P. Conner, "Nonlinear elastic response of shocked fused silica", presented at the 118th Meeting of the Acoustical Society of America, St. Louis, MO (November 1989).
27. J.B. Aidun and Y.M. Gupta, "Characterizing shock-induced, high-pressure phases", presented at the March 1990 APS Meeting, Anaheim, CA (March 1990).

APPENDIX B
Personnel and Equipment Support

Research Personnel

1. P.M. Bellamy, Project Engineer, (partial support), Shock Physics
2. S.C. Jones, Visiting Assistant Professor, Laser Physics
3. H. Lueck, Research Physicist, Laser Physics
4. X.-A. Shen, Postdoctoral Research Associate, Laser Physics
5. J.L. Thompson, Engineering Technician, (partial support), Shock Physics

Graduate Students

1. J.B. Aidun, Ph.D. Student, Shock Physics
2. R.T. Casper, Ph.D. Student, Laser Physics
3. M.P. Conner, M.S. Student, Shock Physics
4. X.-A. Shen, Ph.D. Student, Laser Physics
5. L. Simpson, M.S. Student, Laser Physics
6. B.S. Stapleton, M.S. Student, Shock Physics

Equipment Acquisition

1. Imacon 790 Streak Camera
2. Dual Beam Oscilloscope
3. CCD Based Multichannel Analyzer
4. Triplemate Spectrograph and Gratings
5. Argon-Ion Pumped CW Dye Laser
6. Fast Gate for EG&G OMA III
7. Raman Shifter

8. Joulemeter Display
9. D-1000 Pre-Amp Remote Control Mount and Interface
10. Microcomputer, Keyboard and Hard Disk
11. Optical Mounts and Laser Accessories

APPENDIX C
Workshop on
Nonlinear Material Response to Very Rapid Energy Deposition
at Washington State University
Department of Physics
Pullman, WA 99164
May 17, 18, 1989

Wednesday, May 17

8:20: Opening Remarks by Dr. S. Wu, Program Manager, AFOSR

Shock Deformation in Solids

8:30: Y.M. Gupta, "Shock Wave Measurements in Geophysical and Ceramic Materials: Limitations and Needs"

9:00: M.P. Conner, "Shear Wave Measurements to Determine the Nonlinear Elastic Response of Fused Silica Under Shock Loading"

9:30: J.B. Aidun, "Shear Wave Measurements to Characterize Shock Induced Phase Transformations in Polycrystalline Calcite"

10:00: Break

10:15: X.A. Shen, "Effect of Crystal Orientation on R-Line Luminescence Measurements in Shocked Ruby"

10:45: S.M. Sharma, "Modeling of R-Line Luminescence to Understand Shock Deformation in Ruby Crystals"

11:15: Discussion

12:00: No Host Luncheon at the Compton Union Building

Laser Damage of Optical Materials

1:30: P.F. Braunlich, "Historical Review of Laser Damage Studies: Theories, Methods, Controversies"

2:00: S.C. Jones, "Models for and Measurements of Multiphoton Initiated Prebreakdown Energy Absorption in Wide Gap Insulators"

Laser Damage of Optical Materials — Continued

- 2:30: X.A. Shen, "Multiphoton Absorption, Lattice Heating and Laser Damage in Alkali Halides"
- 3:00: Break
- 3:15: R.T. Casper, "Models and Measurements for Single- and Multi-pulse Laser Damage in Alkali Halides"
- 3:45: Discussion

Thursday, May 18

Invited Lectures:

- 8:15: J.W. Shaner, "Soft Sphere Modeling of Fluid Phase Thermodynamics: Applications to Rapid Energy Deposition"
- 9:05: Break
- 9:25: B.S. Holmes, "An Overview of Recent Laser Effects Studies at SRI"
- 10:15: Discussion and Closing Remarks
- 10:45: Tour of the Shock Dynamics Laboratory and the Laser Physics Laboratory.

Nonlinear Material Response to Very Rapid Energy Deposition

Department of Physics
Washington State University
Pullman, WA 99164-2814

May 17, 18, 1989

John Aidun
Department of Physics
Washington State University
Pullman, WA 99164-2814

Kent Anderson
AFWL/NTES
Kirtland AFB
Albuquerque, NM

Paul Bellamy
Department Physics
Washington State University
Pullman, WA 99164-2814

Peter F. Braunlich
Department of Physics
Washington State University
Pullman, WA 99164-2814

Tom Casper
Department of Physics
Washington State University
Pullman, WA 99164-2814

William Cook
AFATL/MNW
Eglin AFB, FL 32542

J.L. Ding
MME Department
Washington State University
Pullman, WA 99164-2920

George E. Duvall
Department of Physics
Washington state University
Pullman, WA 99164-2814

Jiawen Fan
Department of Physics
Washington State University
Pullman, WA 99164-2814

Carl Frederickson
Department of Physics
Washington State University
Pullman, WA 99164-2814

John Gill
AFWL/NTESG
Kirtland AFB
Albuquerque, NM 87117-6008

Y.M. Gupta
Department of Physics
Washington State University
Pullman, WA 99164-2814

Rick Gustavsen
Department of Physics
Washington State University
Pullman, WA 99164-2814

Bayard S. Holmes
SRI International
333 Ravenswood Avenue
Menlo Park, CA 94025

Scott C. Jones
Department of Physics
Washington State University
Pullman, WA 99164-2814

John Kenemuth
AFWL/AROD
Kirtland AFB
Albuquerque, NM 87117-6008

Horst Lueck
Department of Physics
Washington State University
Pullman, WA 99164-2814

Phil Marston
Department of Physics
Washington State University
Pullman, WA 99164-2814

Constantina Poga
Department of Physics
Washington State University
Pullman, WA 99164-2814

John Shaner
Los Alamos National Laboratories
P.O. Box 1663, MS J970
Los Alamos, NM 87545

Surinder S. Sharma
Department of Physics
Washington State University
Pullman, WA 99164-2814

Xiao-An Shen
Department of Physics
Washington State University
Pullman, WA 99164-2814

Mike Winey
Department of Physics
Washington State University
Pullman, WA 99164-2814

Mike Wong
Department of Physics
Washington State University
Pullman, WA 99164-2814

Spencer Wu
AFOSR/NA
Bolling AFB, DC 20332

Hussein Zbib
MME Department
Washington State University
Pullman, WA 99164-2920